Supporting Information

Table S1. Onset transition temperatures used to calculate reduced temperatures, T-T_{NI}, of solutions of 5 wt % or 10 wt % coil/SGLCP diblock copolymers dissolved in 5CB or d_{19}5CB.

	Polymer	5 wt %		10 wt %	
		T_{NI} in 5CB [°C]	T_{NI} in d_{19}5CB [°C]	T_{NI} in 5CB [°C]	T_{NI} in d_{19}5CB [°C]
End-On	470(40)ABSiCB4	35.4	35.2	37.3	36.9
	390(60)ABSiCB4	35.4	33.4	35.3	33.3
	420(80)ABSiCB4	35.7	34.5	36.4	35.3
	320(120)ABSiCB4	34.6	33.0	35.0	32.9
Side-On	590(40)ABSiBB	35.5	32.1	35.5	33.0
	530(60)ABSiBB	36.0	32.5	36.2	33.5
	620(80)ABSiBB	35.5	32.5	35.5	33.5
	450(120)ABSiBB	35.8	33.0	36.0	33.5

Results from Side-On SGLCP's

The SANS and rheology experiments reported in the main text were also performed on a set of coil/SGLCP diblocks having mesogenic side-groups attached "side-on", with the mesogen's long axis parallel to the polymer backbone (Figure S1). Side-on and end-on polymers were synthesized from the same set of four PS-PB diblocks and one PB homopolymer using methods described in Reference 36. Side-on polymer properties are summarized in Table S2.

All experimental methods for side-on and end-on polymers were identical except the percentage of reacted 1,2-butadiene monomers in side-on polymers was calculated from the relative intensities of peaks at δ = 4.9 ppm (characteristic of vinylic hydrogens in 1,2-butadiene) and δ = 3.6 ppm (characteristic of hydrogens in the side-on mesogen's alkyl tails). Also, rheological temperature ramps on side-on solutions were performed at a rate of 0.5 °C/min, instead of the rate of 1 °C/min used for end-on polymers. TEM experiments were not performed using side-on polymers.

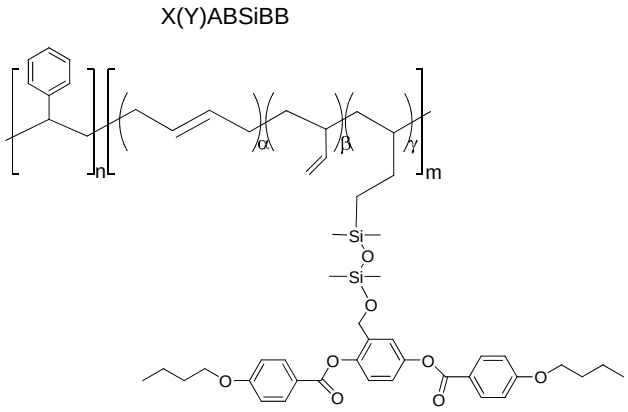


Figure S1. Chemical structure of the side-on side-group liquid crystal polymers. Diblock copolymers have a coil block composed of polystyrene and the SGLCP homopolymer has $n = 0$. The polymers' names are derived from the molecular weights of the liquid crystal polymer block, *X*, and the coil block, *Y*, in units of kg/mol, the letters "AB" or "H" to indicate a diblock copolymer or homopolymer, respectively. In addition to monomers having an attached mesogen, the polymers also contain some residual 1,2- and 1,4-butadiene monomers. Compositions, expressed as the mole fractions α,β, and γ, are given in Table S2.

Table S2. Molecular weight, conversion, and polydispersity of the side-on liquid crystalline polymers.

Name	PS Block M_n [kg/mol]	SGLCP Block				PDI[a]
		M_n [kg/mol]	Mole Fraction 1,2 PB	Mole Fraction 1,4 PB	Mole Fraction LC	
500HSiBB	-	497	0.07	0.11	0.82	1.15
590(40)ABSiBB	43	592	0.21	0.01	0.78	1.37
530(60)ABSiBB	59	534	0.24	0.03	0.73	1.09
620(80)ABSiBB	83	620	0.09	0.05	0.86	1.13
450(120)ABSiBB	121	445	0.31	0.01	0.68	1.65

[a]PDI = Polydispersity Index (M_w/M_n)

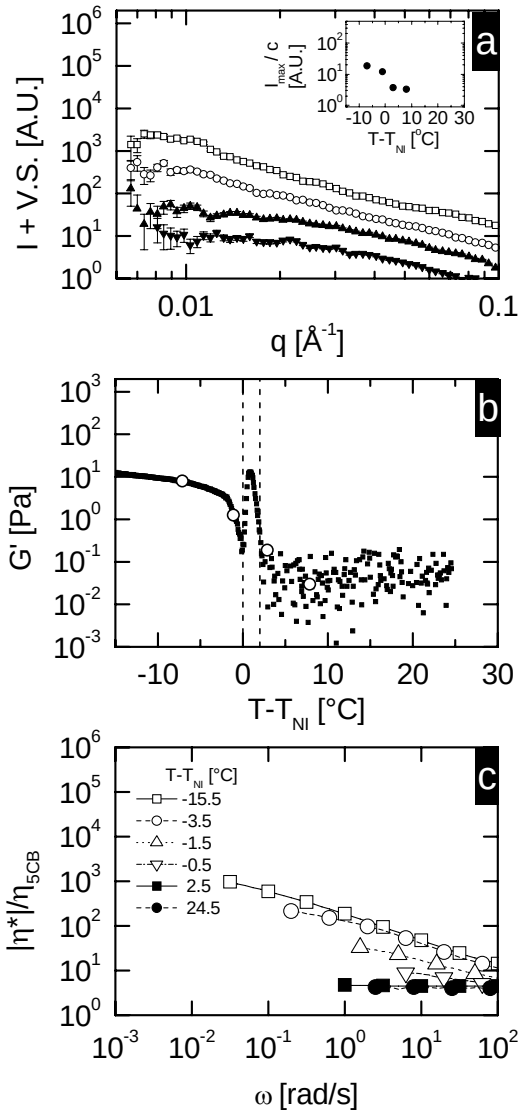


Figure S2. SANS and rheology of 5 wt % 590(40)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at $T - T_{NI} = -7.1$ °C (□), -1.1 °C (○), 2.9 °C (▲), and 7.9 °C (▼). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). Dashed lines indicate the beginning and end of the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

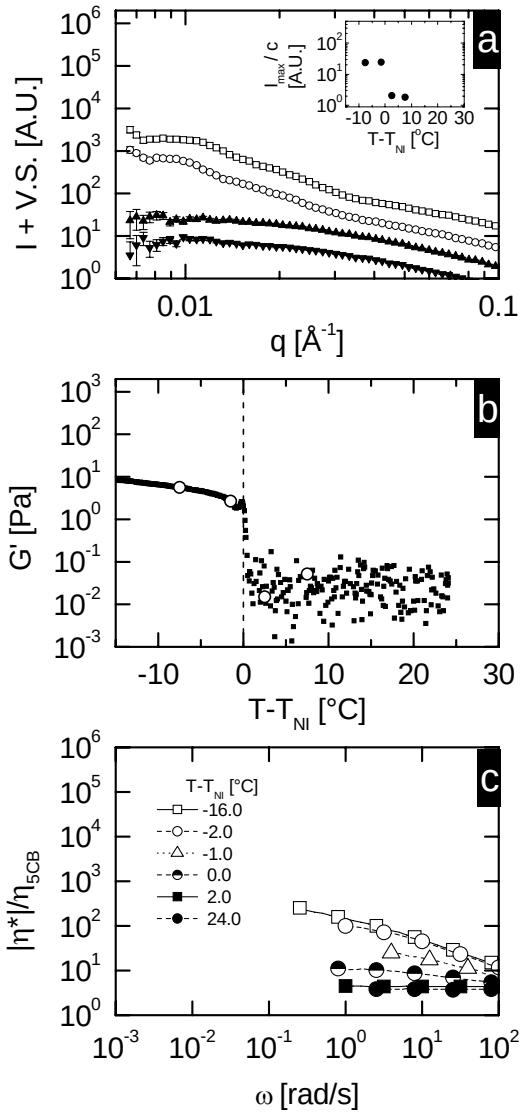


Figure S3. SANS and rheology of 5 wt % 530(60)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at $T - T_{NI} = -7.5$ °C (□), -1.5 °C (○), 2.5 °C (▲), and 7.5 °C (▼). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). A dashed line indicates the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

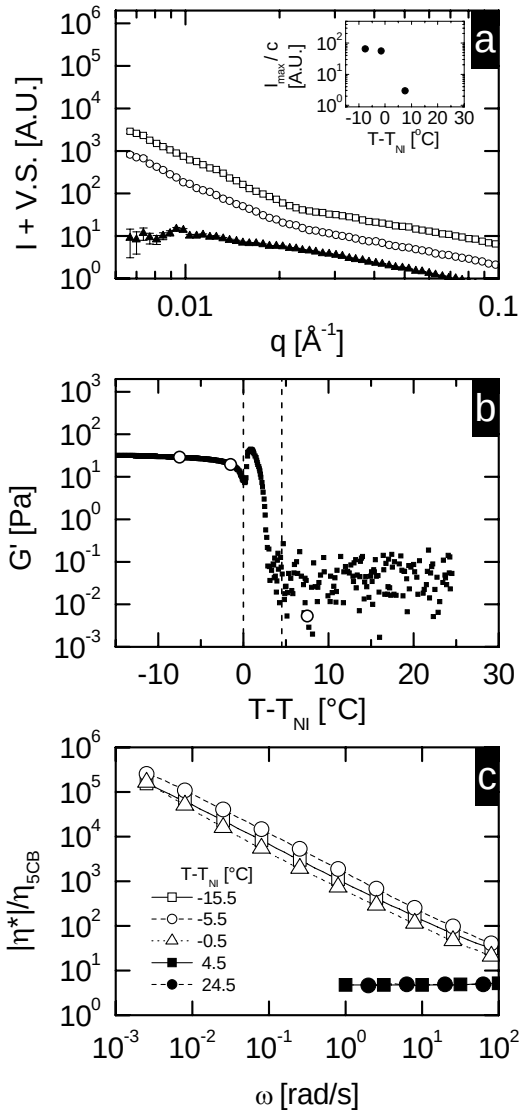


Figure S4. SANS and rheology of 5 wt % 620(80)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at T - T_{NI} = -7.5 °C (□), -1.5 °C (○), and 7.5 °C (▲). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). Dashed lines indicate the beginning and end of the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

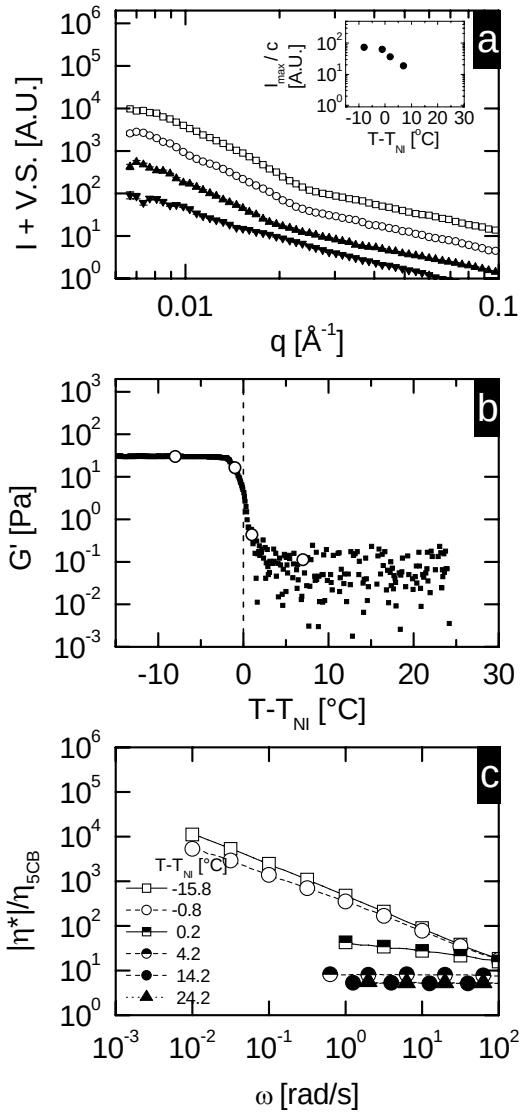


Figure S5. SANS and rheology of 5 wt % 450(120)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at T - T_{NI} = -8.0 °C (□), -1.0 °C (○), 2.0 °C (▲), and 7.0 °C (▼). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). A dashed line indicates the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

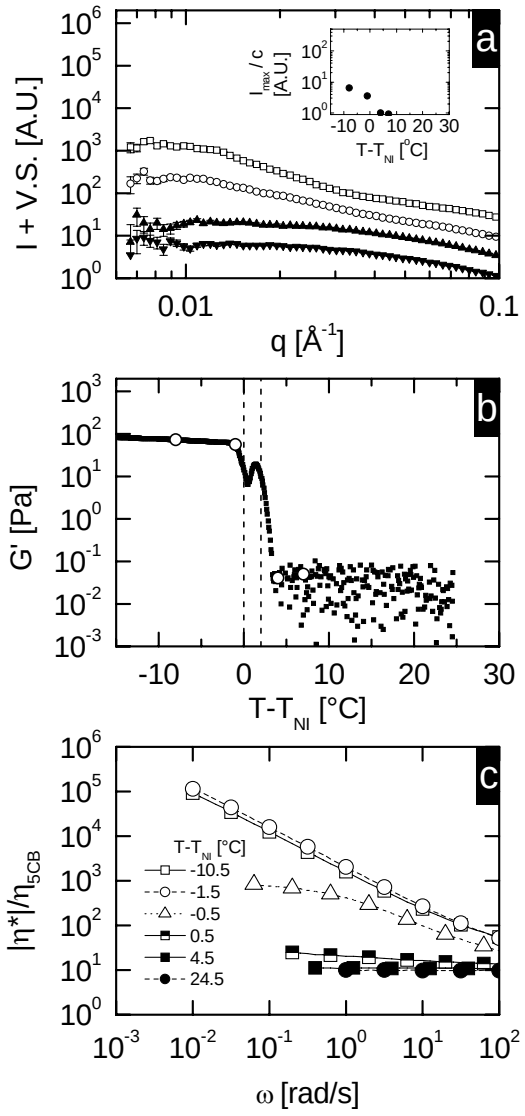


Figure S6. SANS and rheology of 10 wt % 590(40)ABSiBB. (a) The low-*q* portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at $T - T_{NI} = -8.0$ °C (□), -1.0 °C (○), 4.0 °C (▲), and 7.0 °C (▼). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). Dashed lines indicate the beginning and end of the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

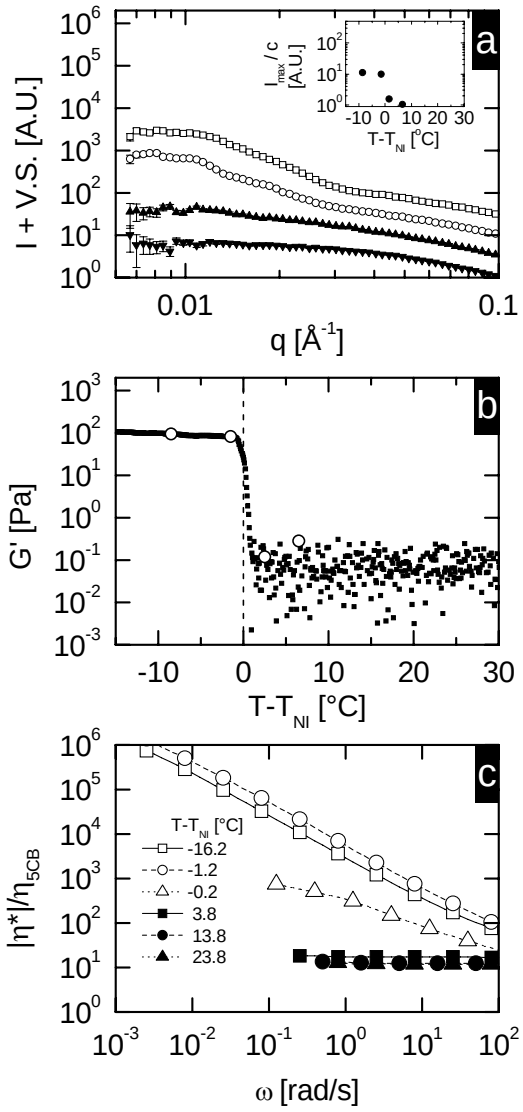


Figure S7. SANS and rheology of 10 wt % 530(60)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at $T - T_{NI} = -8.5$ °C (□), -1.5 °C (○), 1.5 °C (▲), and 6.5 °C (▼). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). A dashed line indicates the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

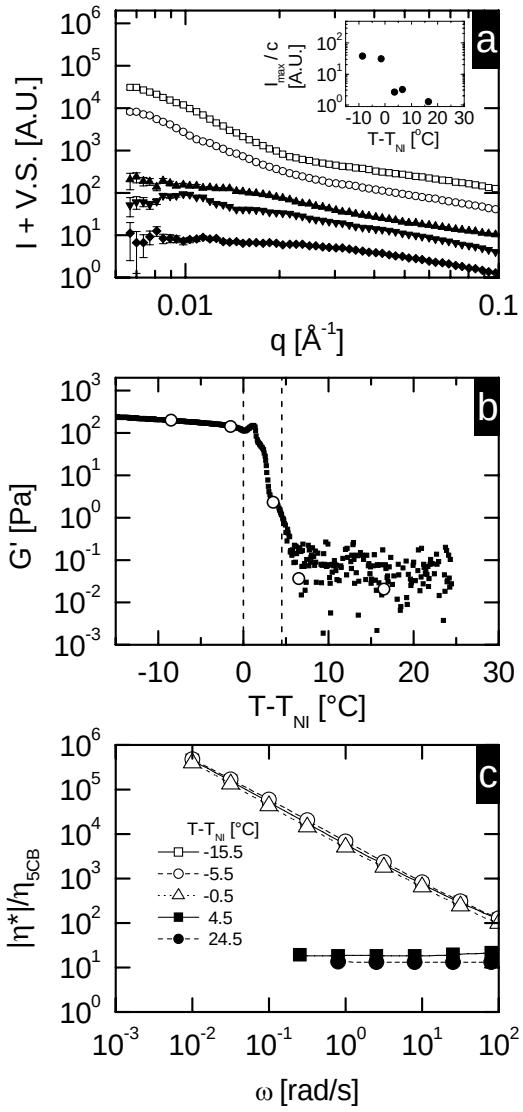


Figure S8. SANS and rheology of 10 wt % 620(80)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at T - T_{NI} = -8.5 °C (□), -1.5 °C (○), 3.5 °C (▲), 6.5 °C (▼), and 16.5 °C (♦). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega = 10$ rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). Dashed lines indicate the beginning and end of the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.

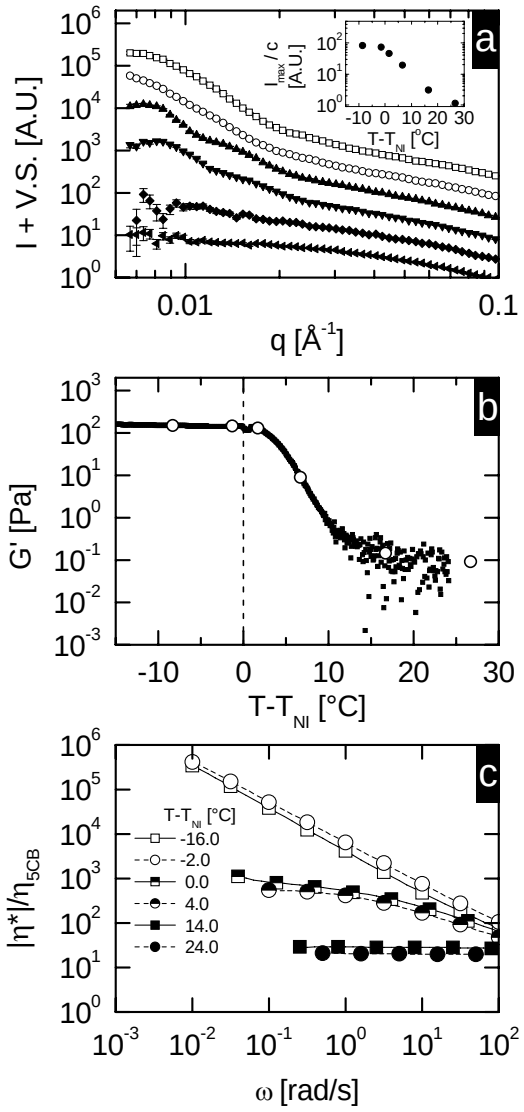
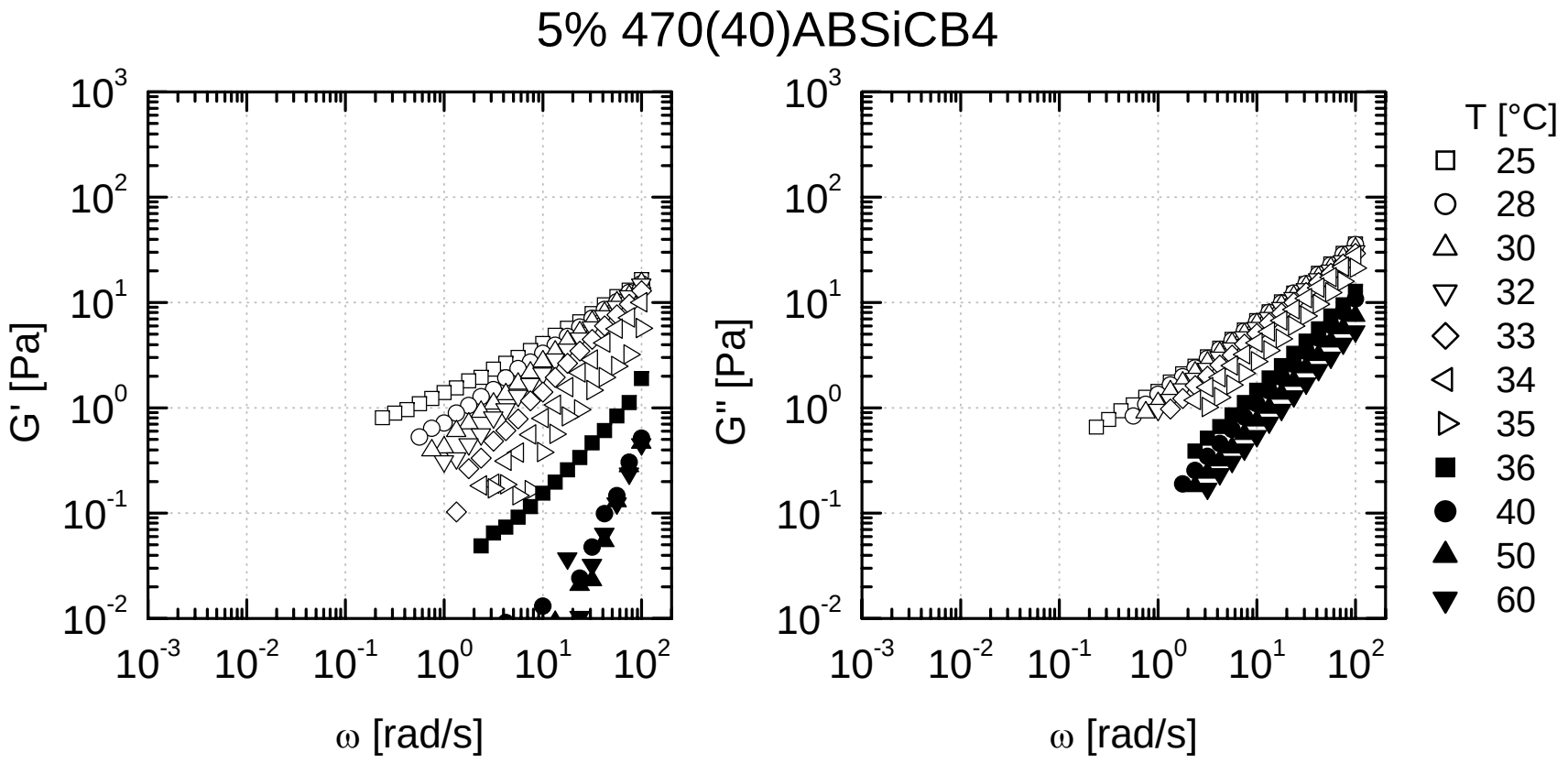
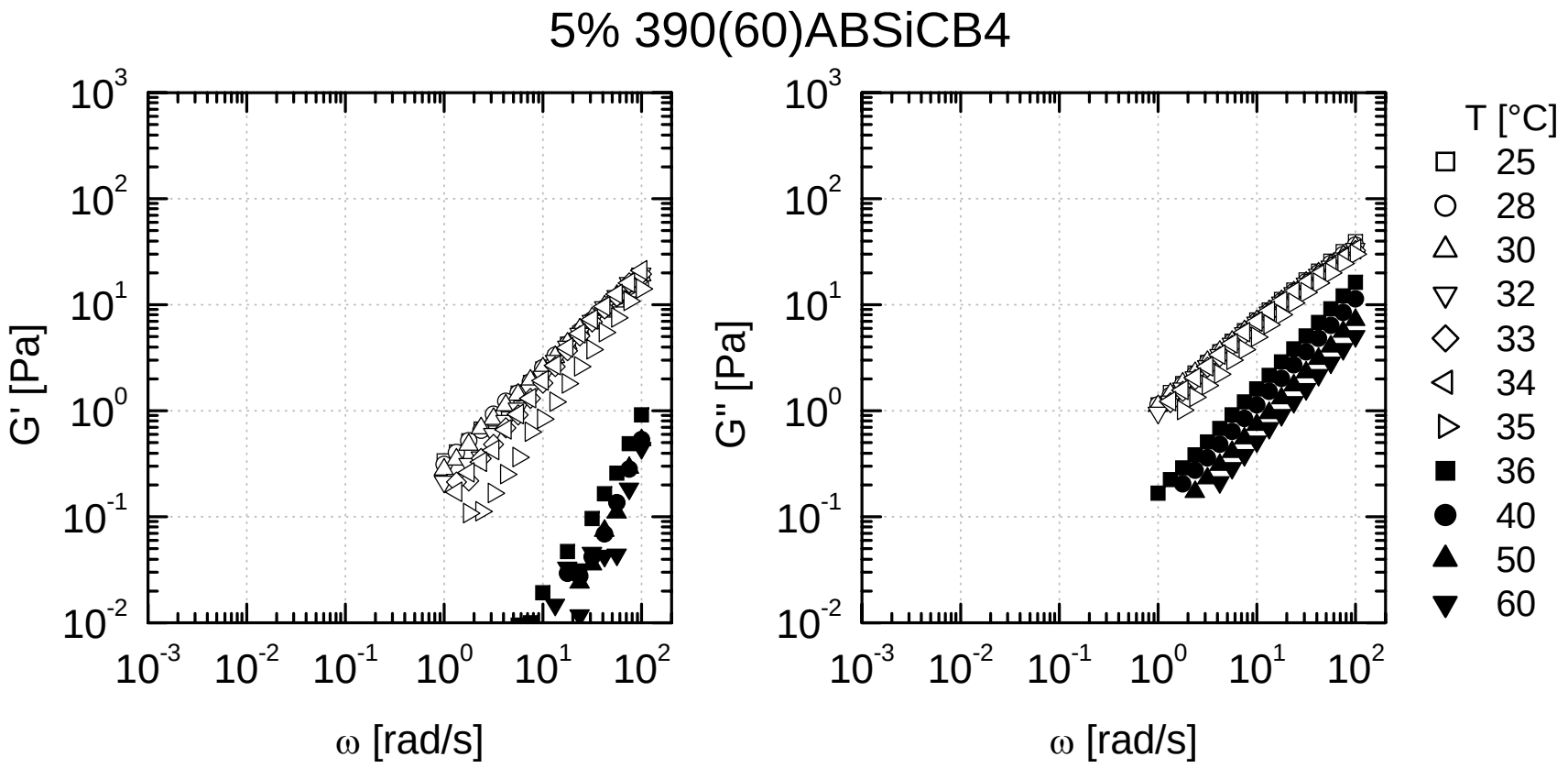


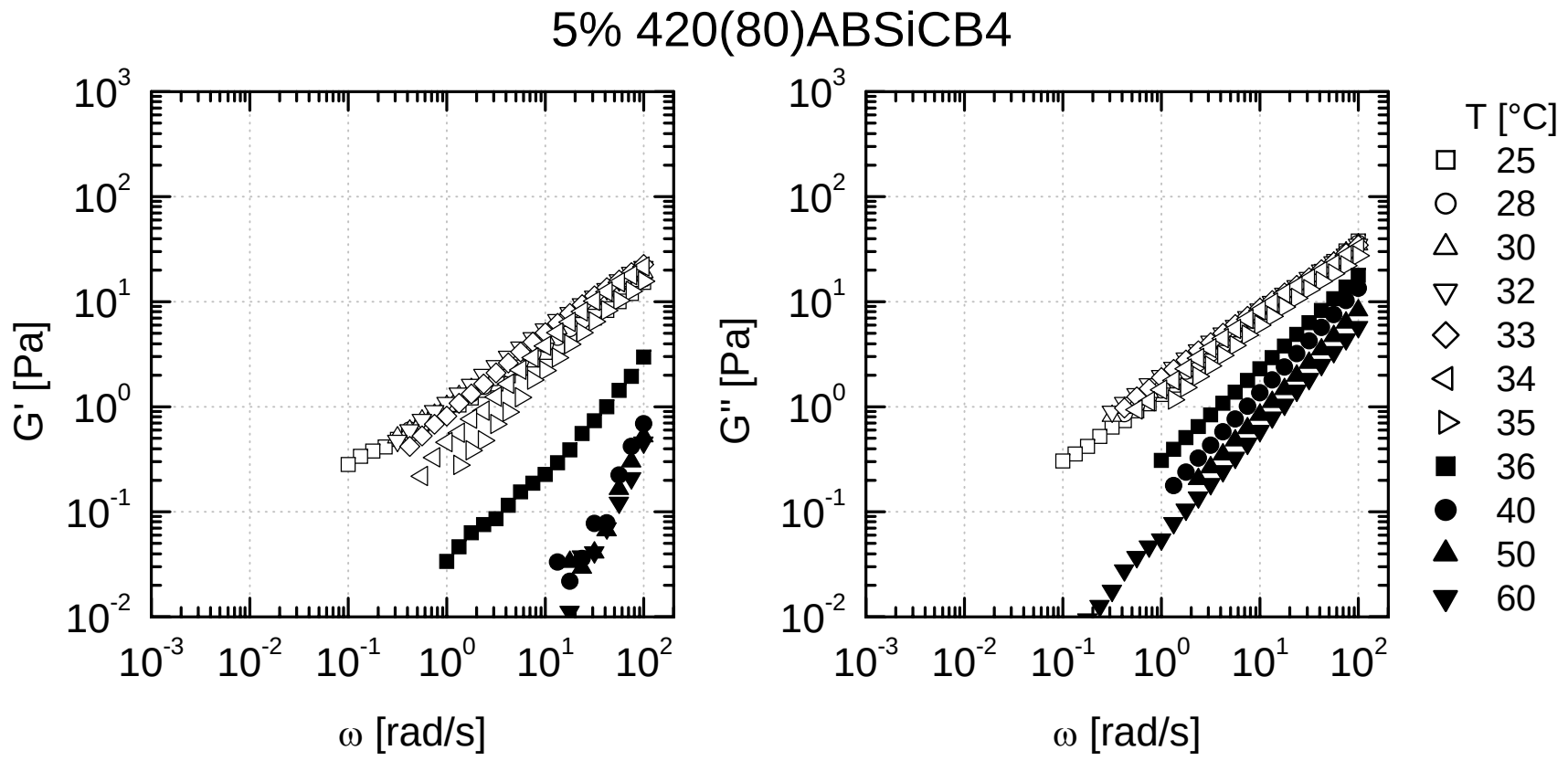
Figure S9. SANS and rheology of 10 wt % 450(120)ABSiBB. (a) The low-q portion of SANS patterns collected in the nematic phase (open symbols) and the isotropic phase (closed symbols) at T - T_{NI} = -8.5 °C (□), -1.5 °C (○), 1.5 °C (▲), 6.5 °C (▼), and 16.5 °C (♦), and 26.5 °C (◄). Patterns are successively shifted upward from the highest temperature by powers of three for clarity. (b) The storage modulus, $G'(\omega$ = 10 rad/s), as a function of temperature upon heating a rate of 0.5 °C/min. White circles indicate the reduced temperatures corresponding to SANS patterns in (a). A dashed line indicates the nematic-isotropic transition. (c) Frequency-dependence of the reduced complex viscosity, $|\eta^*|/\eta_{5CB}$, at various temperatures in the the nematic phase (open symbols) and the isotropic phase (closed or half-filled symbols). In the isotropic phase half-filled symbols are used to indicate viscoelastic fluids while closed symbols indicate viscous liquids.



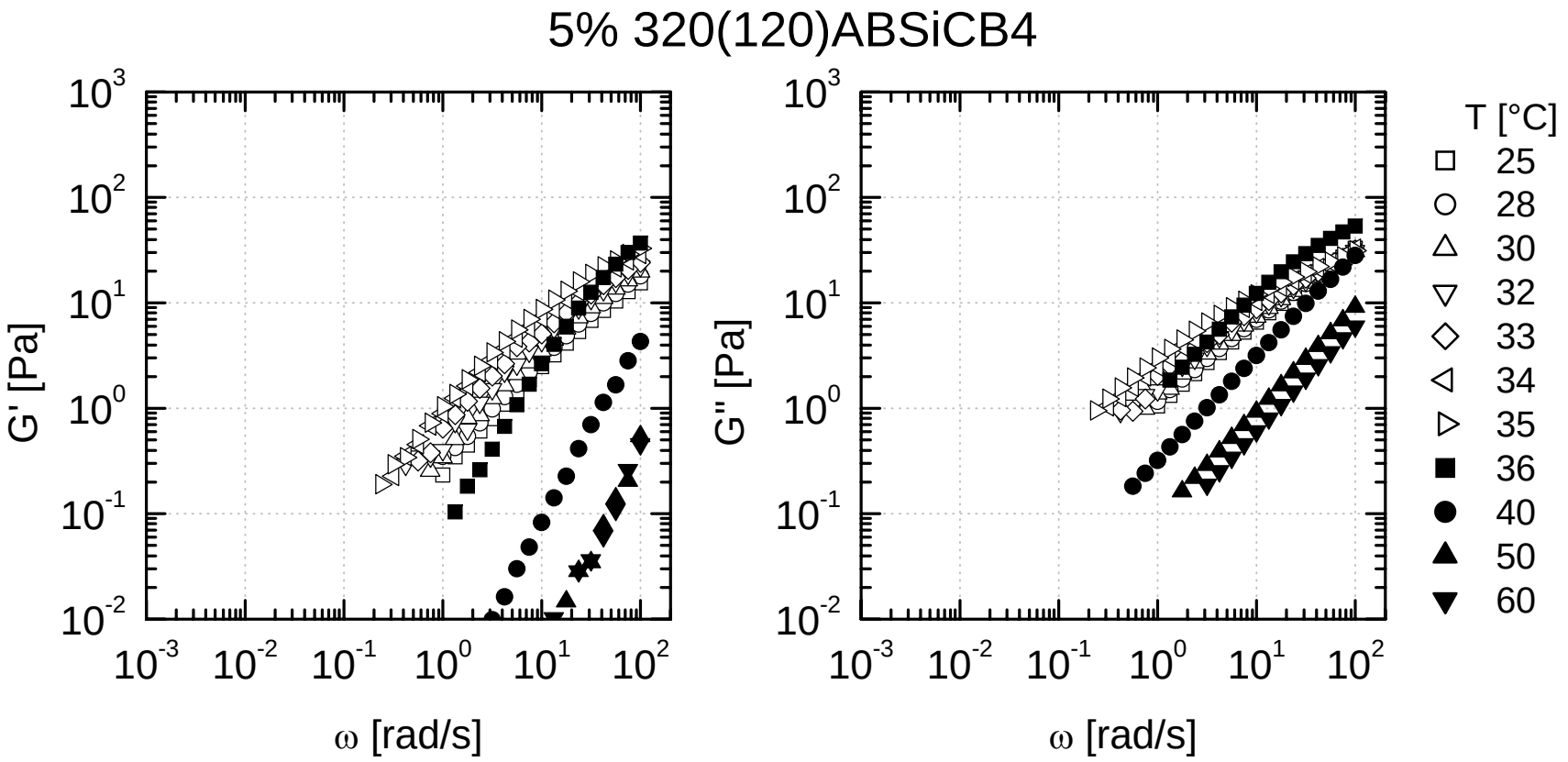
Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 5 wt % 470(40)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



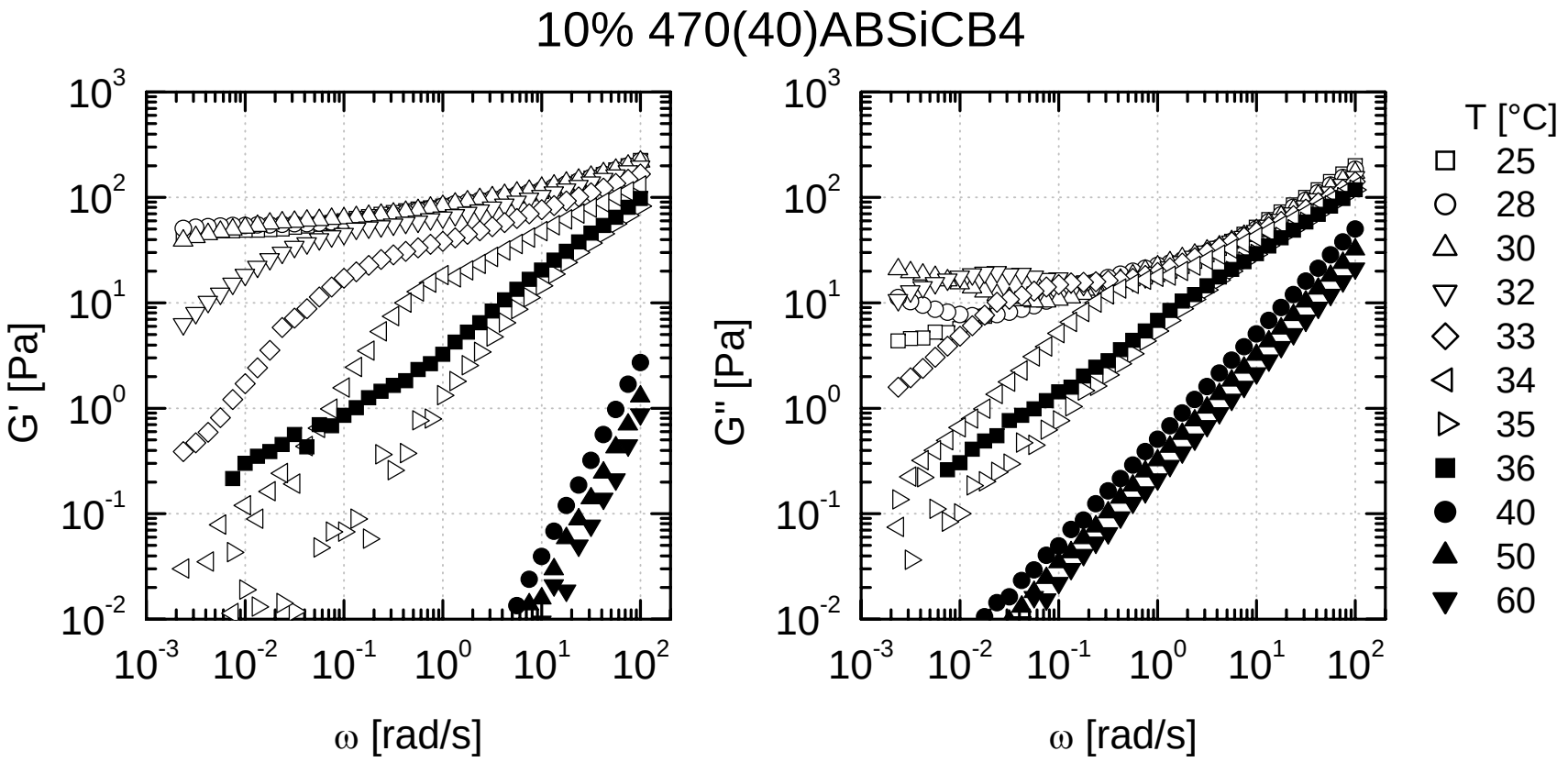
Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 5 wt % 390(60)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



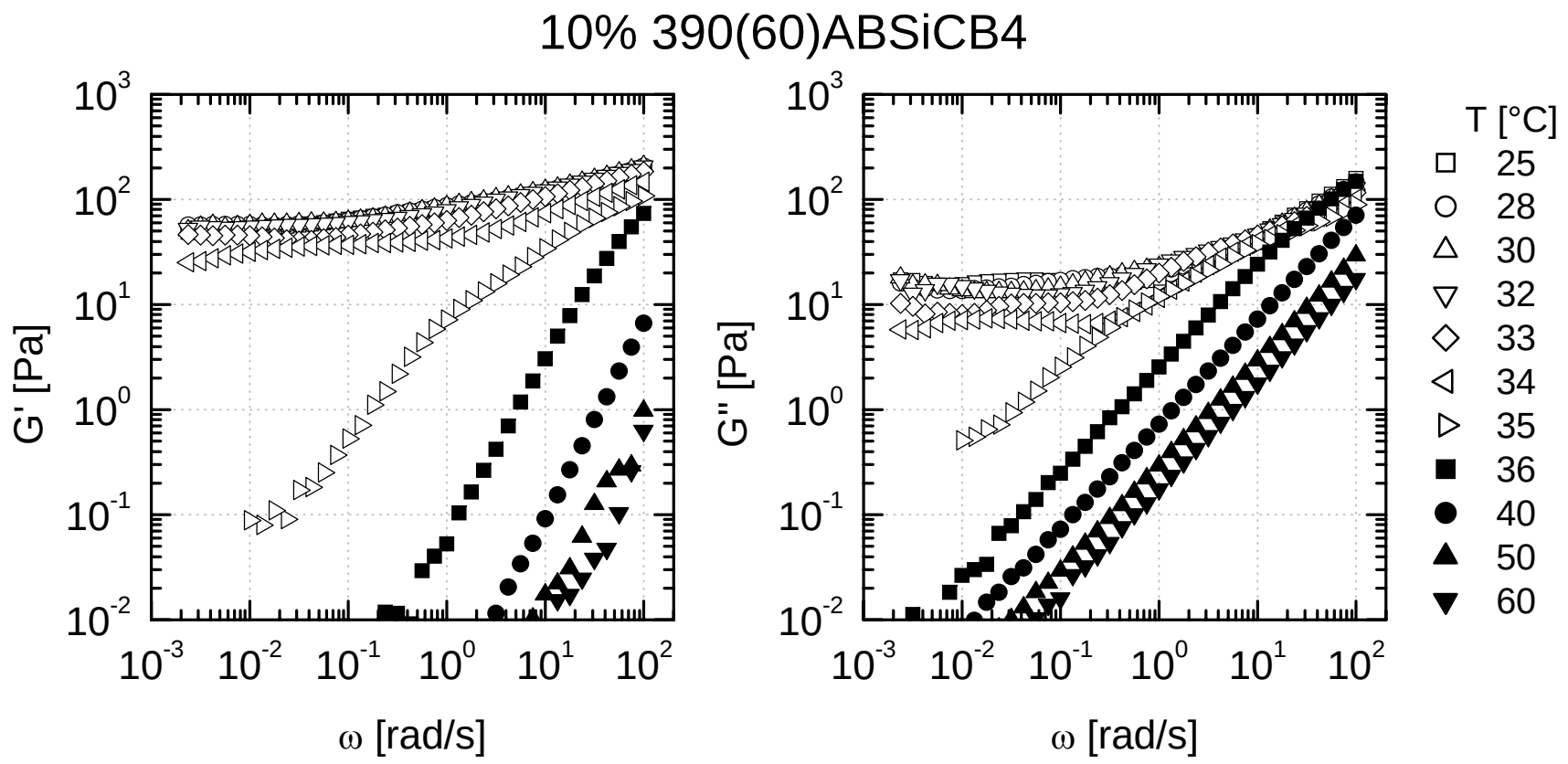
Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 5 wt % 420(80)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



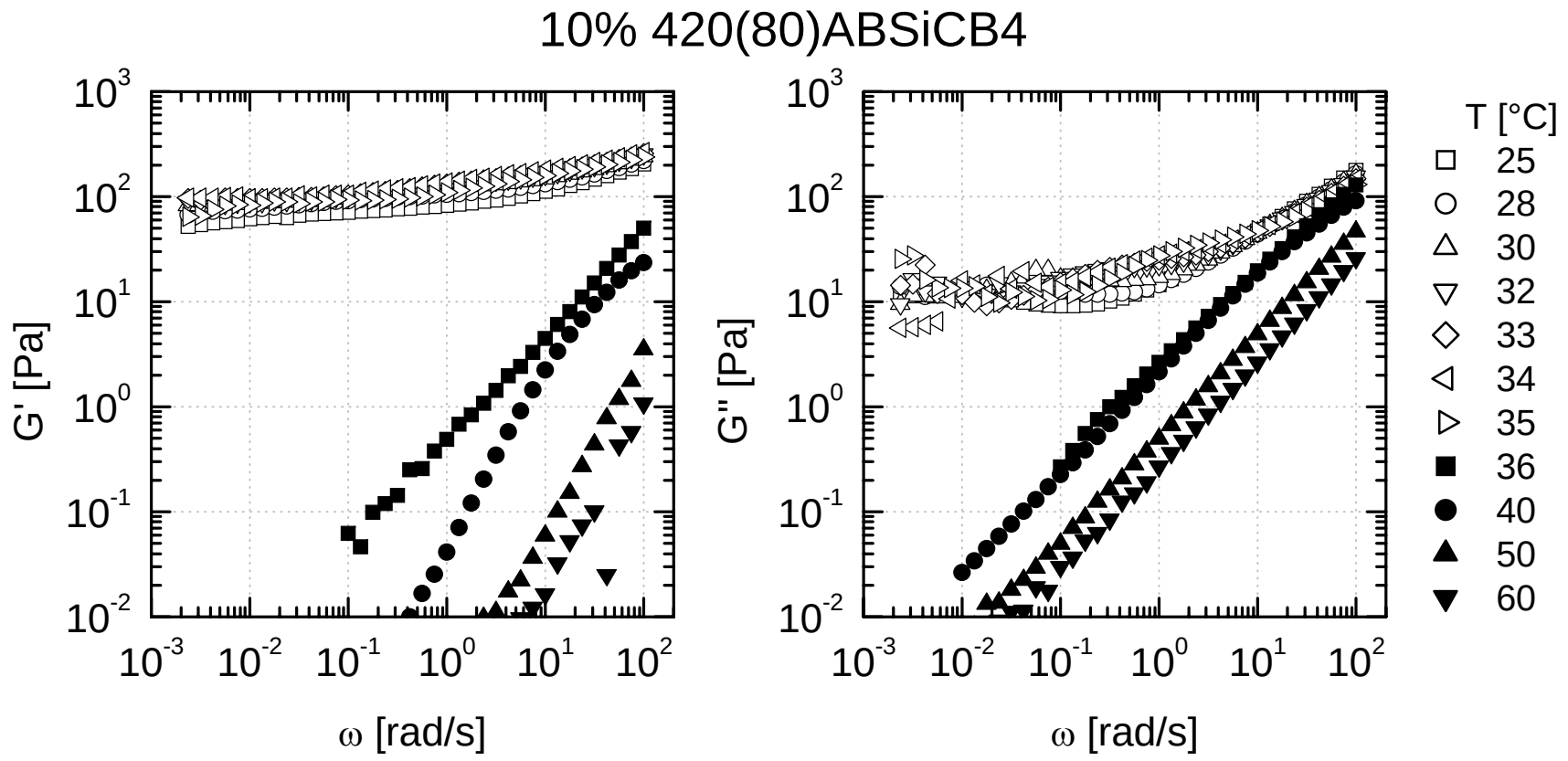
Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 5 wt % 320(120)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



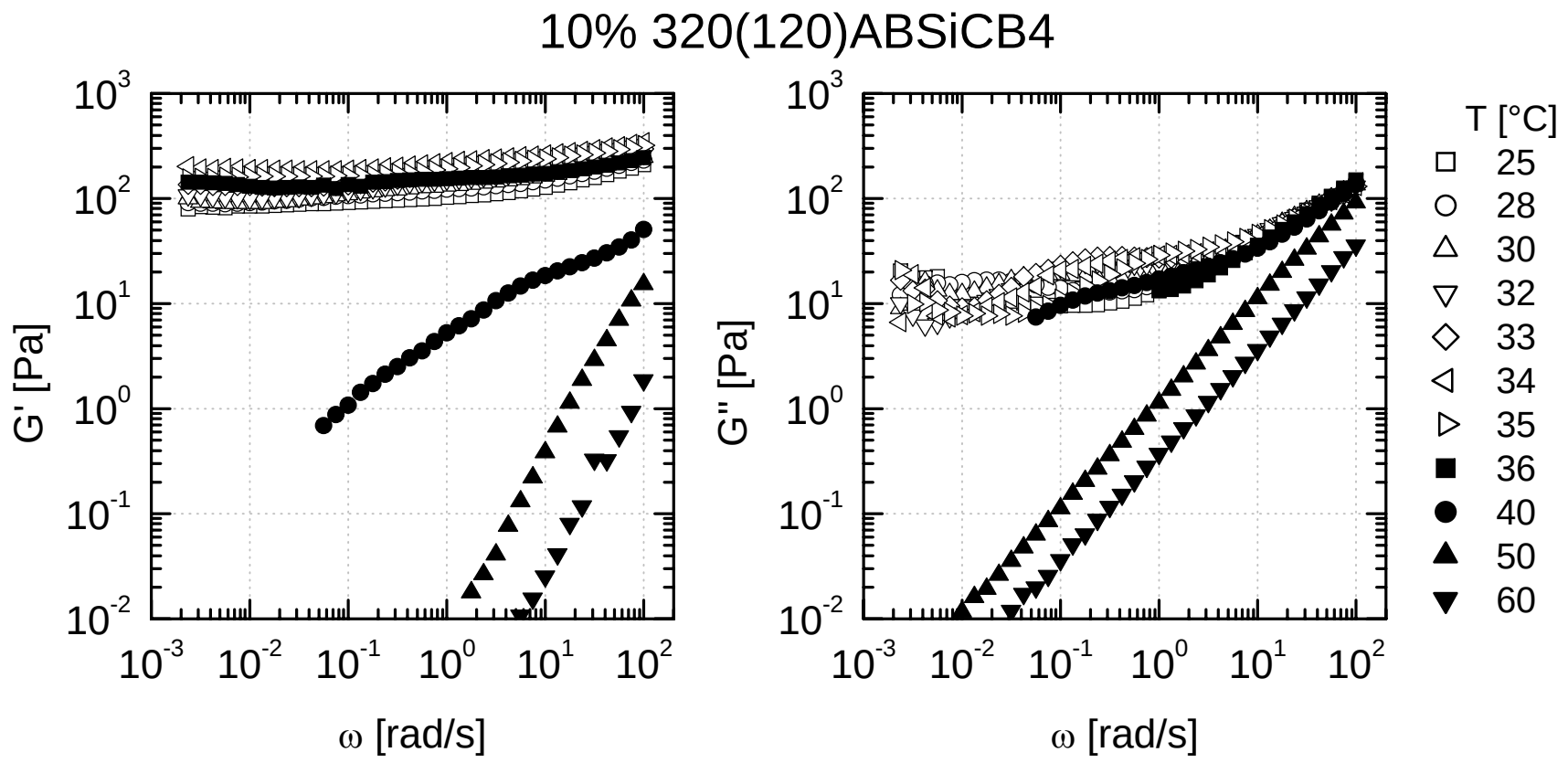
Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 10 wt % 470(40)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 10 wt % 390(60)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 10 wt % 420(80)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).



Frequency (ω) dependence of the storage (G') and loss (G'') modulus of 10 wt % 320(120)ABSiCB4 in 5CB as a function of temperature (T) in the nematic phase (open symbols) and the isotropic phase (closed symbols).